Initial Studies of Curium(III) Hydrolysis Using Time-Resolved Laser Fluorescence Spectroscopy

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Time-resolved laser fluorescence spectroscopy (TRLFS) has been used to study many inorganic complexes. In actinide science, TRLFS has been employed to provide information on the complexation of hexavalent uranium and trivalent curium compounds. Quantitative analysis of curium compounds has proven successful at very dilute concentrations, reported as low as 1 x 10^{-9} M[1]. Due to curium's strong f-f electronic transition and subsequent vibronic relaxation, curium compounds have relatively simple spectra that are easy to interpret. These attributes have made TRLFS a very practical choice in the study of various curium compounds.

Our initial study focuses on the hydrolysis of curium and its relationship to the fluorescence lifetime,τ. The fluorescence lifetime for curium(III) is defined as the average amount of time it takes for an excited atom to return to its ground state. A lifetime is representative of both radiative and nonradiative forms of decay, which in turn gives description about the ion's complexation environment.

Curium solutions (10^{-6}M) were prepared for comparison of emission spectra. Spectra were obtained throughout November and December. As shown in figure 1, the free curium emission spectrum is characterized by a single peak centered at 594 nm.

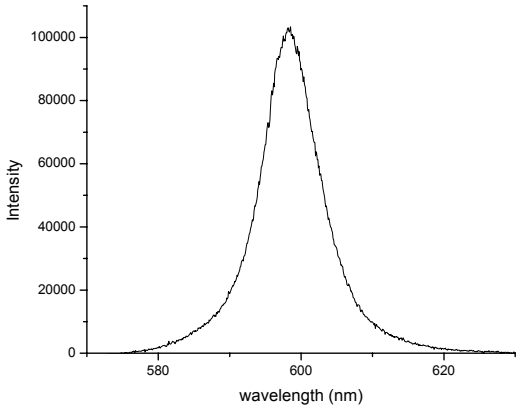

FIG. 1:The emission spectrum of curium(III) (1*10^{-6}M) in 0.1M $HClO_4$

The decay of curium's fluorescence was also studied, as shown in figure 2. Uncomplexed curium's lifetime was found to be 61 microseconds. This is in agreement with the literature[2]. Results have already shown the formation of two distinct hydrolysis products, as evidenced by longer lifetimes and peak shifts, the latter shown in figure 3. This is in agreement with previous experiments[3].

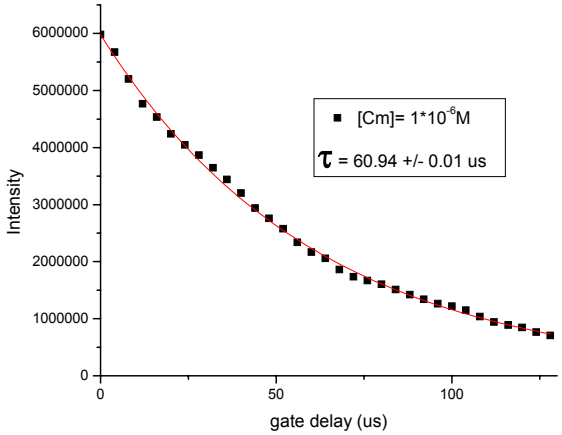

FIG. 2: A time-resolved study of curium (III) (1*10^{-6}M) in 0.1M $HClO_4$

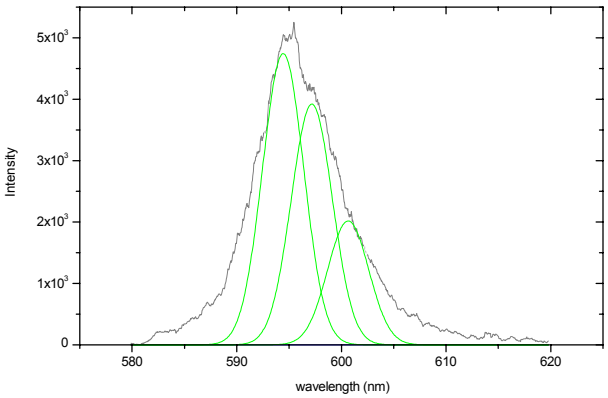

FIG. 3: Hydrolysis products at pH=6.45. [Cm]=850 nM in 0.1M $NaClO_4$

Various carboxylic acid solutions (propionic, butyric and valeric acids), will be prepared for analysis with TRLFS to determine lifetimes for calculation of complex stability constants. Environmental factors, such as pH and ionic strength, will also be varied to further our understanding of a given model system.

REFERENCES

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